



Bench-scale gasification of cedar wood – Part I: Effect of operational conditions on product gas characteristics

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H I G H L I G H T S

- ▶ The paper presents experimental results on cedar wood gasification.
- ▶ Tests have been conducted under various operating conditions in a bench-scale facility.
- ▶ Thermodynamic equilibrium calculations were performed to predict the equilibrium gas composition.

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A B S T R A C T

The present study was conducted within the framework of R&D activities on the development of gasification and reforming technologies for energy and chemical recovery from biomass resources. Gasification of the Japanese cedar wood has been investigated under various operating conditions in a bench-scale externally heated updraft gasifier; this was followed by thermal reforming. Parametric tests by varying the residence times, gasification temperatures, equivalence ratios (ERs) and steam-to-carbon (S/C) ratios were performed to determine their effects on the product gas characteristics. Thermodynamic equilibrium calculations were performed to predict the equilibrium gas composition and compared with the experimental value.

We found that the product gas characteristics in terms of the H₂/CO ratio, CO₂/CO ratio, and CH₄ and lighter hydrocarbons concentrations are significantly affected by the operating conditions used. Increasing the residence time decreased the CO₂/CO ratio; however, a nominal effect was noticed on H₂ concentration as a function of the residence time. At sufficient residence time, increasing the temperature led to higher H₂ yields, CO efficiency and higher heating value (HHV) of the product gas. The presence of steam during gasification effectively enhanced the proportion of H₂ in the product gas. However, higher S/C ratio reduced the HHV of the product gas. Increasing the ER from 0 to 0.3 increased the H₂ yields and CO efficiency and decreased the HHV of the product gas.

The evolution of CH₄ and lighter hydrocarbons at low gasification temperatures was relatively higher than that at high temperature gasification. The evolution of CH₄ and lighter hydrocarbons at high gasification temperatures hardly varied over the investigated operating conditions.

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1. Introduction

There has been a surge of interest in renewable sources of energy as a result of the dwindling reserves of fossil fuels as well as the emergence of global warming (Balat and Balat, 2009). Japan's energy policy is based on market principles. Nevertheless, it seeks to ensure a stable supply of and the environmentally production and consumption of energy. Currently, oil accounts for about 50% of Japan's total primary energy supply and almost all of it is

imported (Japan Ministry of Economy, Trade and Industry, 2008). Therefore, the government has been promoting nuclear energy as a means to diversify its energy sources. However, the recent Fukushima Daichii nuclear plant crisis caused by the March 11th earthquake and tsunami has posed a serious challenge to the nation's energy security. The nuclear crisis may also complicate Japan's efforts to address climate change challenges. Nuclear energy has been seen as an integral part of Japanese plans to achieve a 25% emission reduction target of greenhouse gases (GHGs) by 2020 (Nakano, 2011).

Gasification of biomass has been viewed as a promising system for producing renewable hydrogen; such a process is beneficial for

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the exploitation of biomass resources and for developing a highly efficient, clean method of large-scale hydrogen production (Wang et al., 2008; Kawamoto et al., 2009).

Owing to the abundant forest resources in Japan, woody biomass can act as a potential candidate to cover part of the country's demand of energy. Furthermore, the limited GHG emissions associated with the use of biomass for energy production can respond to the growing pressure for the achievement of a better environmental sustainability of power generation processes and advocate a long-term target of reducing the global greenhouse emissions to counter climate change.

Moreover, most of houses in Japan used to be built of wood, and the old ones that are torn down produce about 5 Mt of architectural wood salvage per year (Wu et al., 2006). The reuse and recycling of wood have become a serious problem. Therefore, thermal disposal through gasification can be considered as an appropriate recycling means.

The product gas quality is of a major concern for the development and design of a reliable biomass gasification process. Fundamental understanding of the relation between the product gas quality and the operating conditions is indispensable for the commercialization and sustainability of biomass gasification processes. This paper presents the results of a parametric study on the effect of operational conditions on the product gas characteristics in order to find optimum operating conditions for the process of cedar wood gasification.

2. Experimental

2.1. Biomass fuel

In this study, woody biomass gasification was carried out. The Japanese cedar wood was selected as a model biomass fuel because it is extensively used in forestry plantations in Japan. In addition, it is favored for all types of construction work as well as interior panelling. The wood was crushed and sieved to a particle size of 1–2 mm as feedstock. The result of the ultimate and proximate analysis of the cedar biomass feedstock is provided in [Supplemental Material \(SM\)](#), [Table SM-1](#).

2.2. Experimental facility

A bench scale gasification unit was built to conduct the experiments. A schematic representation is given in [Fig. SM-1](#). The system essentially comprised of two kinds of reactors in series: a primary reactor (gasifier) followed by a secondary reactor (thermal reformer) made of SUS310. The primary reactor was designed as an updraft type gasifier (id 35–100 mm, height (H) 1210 mm), whereas the thermal reformer was a tubular-type reactor (id 48 mm, H 1620 mm). All components of the setup were connected by 2.54-cm pipes. The pipe connections were mainly Swagelok straight unions. Both reactors were flanged-pipes and a stainless steel metal gasket was used for the flange. Gasket paste (a water-soluble adhesive paste) was applied to the both sides of the metal gasket to ensure sufficient sealing performance.

Solid products (char, ash) that might be carried by the raw gas were removed from the raw gas by a means of a cyclone. A set of thermocouples were placed at different positions within the whole system to monitor and control the raw and product gas temperatures. A data logger (Keyence) was used for data capture.

The reactors were preheated externally by electric heaters. A temperature controller consol (Koyo Thermo Systems) was used to program and adjust the temperature of the reactors as needed. A set of mantle and ribbon heaters were used to keep the temperature of pipes, fittings, sampling ports, and cyclone at a constant

temperature of 473 K. Exposed parts that cannot be heated by mantle or ribbon heaters are kept warm by applying a thick wrap insulation (a fiberglass felt mat called Isowool).

Steam was generated by passing a stream of distilled water through a tube-type furnace (KTF-030N, Koyo Thermo Systems). The water was pumped via a ceramic pump (VSP-2050, EYELA). Nitrogen was used to carry the gasification product gas as well as for purging. Nitrogen was supplied under different flow rates to enable studying the effect of residence time.

The reported results were expressed in terms of nitrogen-free basis. Oxygen was used as a gasifying medium and supplied from the bottom of the gasifier. All the gas flows were controlled with mass flow controllers.

The feedstock fuel was filled in a straight walled cylinder that was carefully sealed. The fuel was fed to the reactor via a circle-type table feeder (Yoshikawa). Wide range of discharge rates were possible by adjusting the variable motor speed. Prior to each experiment, the fuel feed rate was calibrated over a range of motor speeds.

Five Swagelok valves in parallel were installed at the exit pipeline to enable sampling. To prevent clogging in these valves, hot gas filter was installed prior to these valves. The filter was a 2.54-cm union packed with a glass wool that was supported on a metal mesh.

2.3. Sampling procedure

In our study, product gas composition, tar, NH_3 , HCN, HCl, H_2S and COS were quantified. Following is a detailed description about methods and apparatuses used in sampling the product gas. Description about methods and apparatuses used in sampling contaminants level in the product gas will be given elsewhere (Aljbou and Kawamoto, 2012). The sampling layout is given in [Fig. SM-2](#).

2.3.1. Product gas composition

A gas stream was continuously withdrawn through a silicon tube connected to valves 1 and 2. The gas stream was subjected to tar removal unit, moisture removal and fine particles filter unit prior to analysis. The tar removal unit consists of two series of impinger bottles (the first series was connected to valve 1 while the other series was connected to valve 2). The connection between the hot silicon tubing and the impinger bottles was designed accurately to assure the tightness of the joint. Each series comprised three 500 mL impinger bottles. The first and second impinger bottles each contained 200 mL of isopropanol while the third impinger bottle was solvent-free. The impingers were immersed in an ice bath. The moisture was removed by adsorption on silica gel granules filled in a set of four impinger bottles. To ensure complete removal of particles from the product gas, a glass filter with pressure sensor was installed downstream of the impingers.

The tar-free gas was online analyzed by two micro-GC 3000 (Agilent Technologies). The first micro-GC was equipped with molecular sieve column (and was used to measure the concentration of H_2 and N_2 every 2 min. The other micro-GC was equipped with three columns (molecular Sieve, Plot-U, OV-73) and was used to measure the concentration of CO , CO_2 , O_2 , CH_4 , C_2H_2 , C_2H_4 , C_2H_6 , C_3H_6 , C_3H_8 , C_6H_6 , C_7H_8 every 4 min. The two online micro-GC were calibrated prior to each experiment using certified zero and span gases.

2.4. Experiment procedure

Before the day of test, the experimental facility was assembled based on a scheduled plan. The pre-washed gasifier and thermal reformer were assembled then placed in their respective positions. Checking for leaks was performed by first closing the system,

pressurizing to a gage pressure of up to 0.01–0.02 MPa by supplying nitrogen at about 5 L min⁻¹ and applying leak check spray (foam) to all connections and fittings. Gas leak was also confirmed by comparing the supply and outlet nitrogen flow rate.

In the day of experiment, start-up procedures began by heating the system to the desired temperature using the external heaters, with air flowing within the system until a stable temperature profile was obtained. Once the oxygen concentration was stabilized, nitrogen then was supplied. During heating, the outlet gas from gasifier was sent to draft using the bypass line.

During heating the system, the gas sampling devices and apparatuses were constructed as described above. In addition, the steam generating furnace was switched on and its temperature was controlled to give the desired steam temperature of 533 K. This temperature was found experimentally optimum to ensure unified contact time in the gasification zoon.

After ensuring the desired system temperature, the table feeder was connected to the gasifier. Then, nitrogen, oxygen and steam are supplied based on prescribed experimental parameters.

After the system was once again stabilized, the table feeder was turned on and the test begins. Quantitative sampling, as described above, was performed after ensuring that the system had attained a steady temperature and product gas composition. For most of the runs, steady product gas composition was obtained after 30–45 min. Each experimental run was conducted for 3 h. During this period of time, stable conditions were ensured. At the end of experiment, feeder, steam, oxygen, and system heaters were shutdown simultaneously. The system was cooled under nitrogen purging. In the following day, the remaining feedstock was removed from the table feeder and char was recovered from the gasifier and cyclone. Each was weighed and sampled.

2.5. Equilibrium calculation

The maximum achievable yield of gas produced from gasification could be estimated by using equilibrium models, given the fuel composition and the equilibrium temperature based on the assumption that gasification reactions are in thermodynamic equilibrium. In this study, the commercially available ASPEN HYSIS software (Ver. 2006.5, license of Tokyo Gas) was used to predict the gas composition under various operating conditions.

In the equilibrium modeling of the gasification process, it was assumed that the biomass was totally converted into gas thus enabling to neglect char formation. The process was assumed to occur instantaneously at equilibrium with volatile products made of H₂, CO, CO₂, CH₄, and H₂O. Tars were assumed to be negligible in the product gas and were not taken into account in this study.

The ASPEN HYSIS Gibbs reactor model was used for volatile partial oxidation and reforming based on the assumption that volatile reactions follow the Gibbs equilibrium. The composition of the product gas was estimated from the volatile reactions with steam and/or oxygen, minimizing the Gibbs free energy for calculation of the equilibrium. The Peng–Robinson equation of state was selected as a property method to calculate properties such as enthalpy, density, heat duty and temperature.

3. Results and discussion

3.1. Gas composition

3.1.1. Effect of residence time

As described in Section 2 part, the gasifier was designed as up-draft type gasifier. An updraft gasifier is one of the oldest and simplest of all designs. Here, the gasification medium (oxygen, steam) travels upward while the bed of fuel moves downward, and thus

the gas and solids are in countercurrent mode. During operation, the contact region between the two countercurrent streams depends on the applied operating conditions. Due to temperature gradient in gasifier, any change in the contact region is believed to affect the performance of gasification. To ensure a uniform contact between the two countercurrent streams, nitrogen was supplied through the top and bottom of gasifier. As oxygen and steam were supplied from the bottom, nitrogen was supplied from the bottom at a rate enough to achieve a total molar flow rate that was equal to the countercurrent nitrogen molar flow rate from the top of gasifier. In addition, to ensure sufficient contact time, the gasification-thermal reforming performance was examined under different residence times. In the context of this paper, residence time was defined as the length of time that the initial supplied gas resides in both gasifier and thermal reformer. The residence time is altered by changing the nitrogen supply rate.

The effect of residence time was studied at two gasification-thermal reforming temperatures, namely: 923 and 1223 K respectively. Fig. 1 shows the permanent gas composition as a function of residence time. At gasification-thermal reforming temperature of 923 K (Fig. 1a), increasing the residence time increased the concentration of CO and decreased the concentration of CO₂. On the other hand, the concentration of H₂ slightly increased with increasing the residence time. Biomass gasification consists of many concurrent and consecutive reactions. In the presence of oxygen, biomass gasification could be promoted by complete and incomplete combustion, thereby producing H₂, CO, CO₂ and water. Beside the permanent gases, char was produced as a result of the devolatilization reaction of the biomass. The change of composition under oxygen gasification depends on the efficiency of contact between the gasifying medium and the biomass and char. The increase in CO concentration and decrease in CO₂ concentration suggest that CO₂ was consumed as a result of reaction with char according to the Boudouard reaction:



Due to the endothermic nature of the Boudouard reaction, increasing the residence time will minimize the heat transfer limitation and will enhance the heat supply to promote this reaction. The slight increase in hydrogen concentration suggests that part of

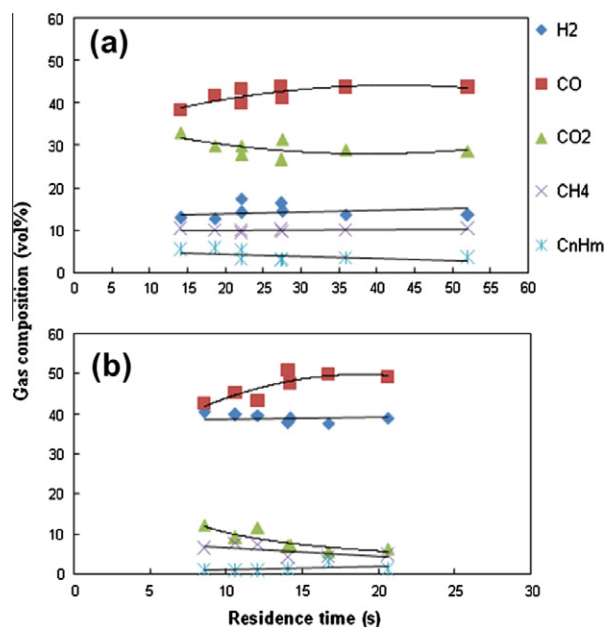


Fig. 1. Gas composition vs. residence time at ER = 0.2, S/C = 0 ((a): temperature = 923 K, (b): temperature = 1223 K).

the produced hydrogen was consumed as a result of reaction with CO. This was evident from the evolution of methane gas in the product gas and it might be expressed as:



This simultaneous production and consumption of hydrogen renders the net concentration of hydrogen almost constant over time.

Increasing the residence time from 14 to 25 s decreased the CO_2/CO ratio from 0.9 to 0.7. On the other hand, any further increase in the residence time did not change the CO_2/CO ratio. The CO_2/CO ratio was almost constant at 0.7. This indicates that equilibrium could be attained when the residence time was 25 s or more.

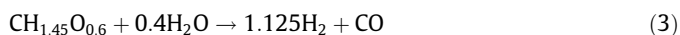
Similarly, at a gasification-thermal reforming temperature of 1223 K (Fig. 1b), increasing the residence time has increased the concentration of CO and decreased the concentration of CO_2 . However, the concentration of H_2 was independent on the residence time. In addition, the concentration of methane decreased with increasing the residence time at this rather high temperature.

At a residence time more than 14 s, both H_2/CO and CO_2/CO ratios were independent on the residence time. This indicates that equilibrium could be attained when the residence time was 14 s or more.

The theoretical and experimental compositions of the product gas at two gasification temperatures are given in Table SM-2. As it can be seen, there was a good agreement between the predicted values and the experimental data at high gasification temperature. However, a remarkable difference can be noticed between the predicated gas composition and the experimental one at low gasification temperature. This difference is attributed to errors associated with the theoretical model assumptions with respect to char formation and tar evolution. In fact, at low gasification temperature, part of the char remains ungasified. This will eventually affect the volatile equilibrium reactions.

3.1.2. Effect of steam-to-carbon ratio (S/C)

The presence of water vapor in the gasifier can assist in the gasification of biomass and the partial oxidation of the hot char. There are different sources for the water vapor in gasifier, such as vapor generated from the solid fuel during pyrolysis or the evaporation of externally-supplied water. Steam gasification of biomass can be expressed as:



where $\text{CH}_{1.45}\text{O}_{0.6}$ is the average formula of the cedar wood based on the actual composition shown in Table S1.

Steam reacts with the hot carbon according to the endothermic water–gas reaction:



In addition, steam can assist in gas reforming through the reduction of steam by carbon monoxide to produce hydrogen. This reaction known as the water–gas shift is expressed as follows:



The effect of S/C ratio was studied at a gasification-thermal reforming temperature of 1123 K. Fig. 2 shows the permanent gas composition as a function of S/C. Increasing the S/C ratio by increasing the steam supply resulted in an increase in the H_2 and CO_2 concentrations and a decrease in the CO concentration. The results show that increasing the S/C ratio from 0 to 2 increased the H_2/CO ratio from 0.7 to 2.6, and the CO_2/CO ratio from 0.4 to 1.5. This might be ascribed to the shift reaction (Eq. (5)). However, excessive increase in the S/C ratio tends to be insignificant to promote further increase in the hydrogen concentration.

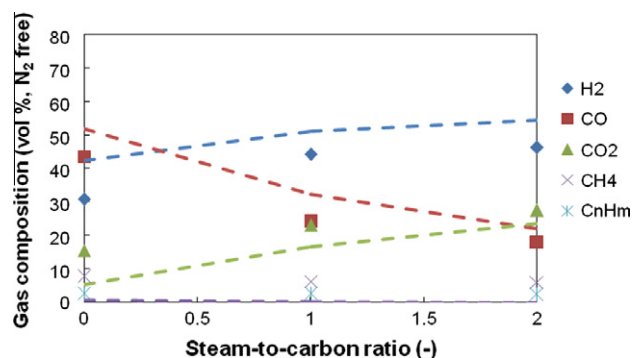


Fig. 2. Gas composition vs. S/C at 1123 K, ER = 0.2 (dash lines represent predicted values).

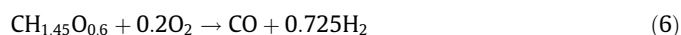
CH_4 and other hydrocarbon concentrations were hardly decreased by increasing the S/C ratio. This might be due to decrease in the contact time caused by the increase of steam supply rate.

The experimental gas composition data have the same trend of the predicted gas composition data with respect to the effect of S/C ratio. Based on the equilibrium calculations, increasing the S/C from 0 to 2 increased the H_2/CO ratio from 0.8 to 2.5, and the CO_2/CO ratio from 0.1 to 1.1.

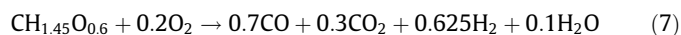
3.1.3. Effect of equivalence ratio (ER)

As it was seen before, the gasification process involved a series of endothermic reactions. The oxidation or combustion of char could assist in providing all the thermal energy needed for the endothermic reactions. Therefore, the addition of oxygen was considered a necessary element in the establishment of a commercial system involving the supply of combustion-based thermal energy to maintain a self-sufficient energy system.

Ideally, it is desired to add the smallest amount of oxygen to convert the biomass into a gas mixture of H_2 and CO according to the formula:



However, there is more energy containing in the CO and H_2 than that in the biomass, so that this reaction would require the addition of energy from an external source. Therefore, some excess oxygen must be added for gasification, producing CO_2 and H_2O as follows:



The effect of the ER on the gas quality was examined for gasification-reforming temperature of 1123 K. Fig. 3 shows the permanent gas composition as a function of ER. An inspection of Fig. 3 shows that the quality of gas obtained from a gasifier depends on the amount of oxygen employed during the gasification. At ER = 0, the product gas had the highest H_2 concentration. H_2 concentration was as high as 50 vol.%. Nevertheless, under this condition, char was excessively produced. Increasing the ER, decreased the H_2 concentration and increased the CO_2 concentration. H_2 was oxidized with oxygen and the more oxygen was added, the more the H_2 declined. Increasing the ER from 0 to 0.3 decreased the H_2/CO ratio from 2.1 to 1.8, whereas the CO_2/CO ratio increased from 0.7 to 1.1. The amount of light hydrocarbons was almost constant with increasing the ER.

With respect to the effect of ER, the experimental gas composition data have the same trend of the predicted gas composition data. However, equilibrium calculations indicated that increasing the ER from 0 to 0.2 had no effect on the H_2/CO ratio. Within this range of ERs, the H_2/CO ratio was constant at 1.6. On the other hand, the CO_2/CO ratio increased from 0.3 to 0.7.

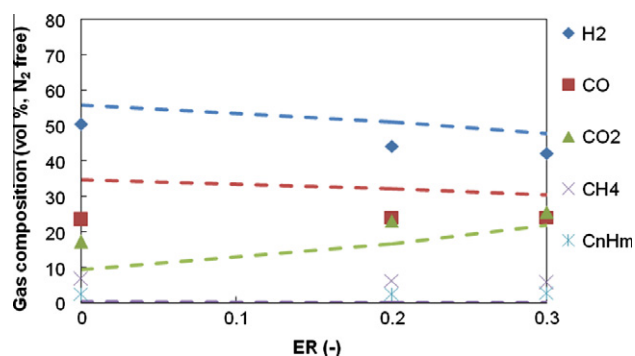


Fig. 3. Gas composition vs. ER at 1123 K, S/C = 1 (dash lines represent predicted values).

3.1.4. Effect of gasification-thermal reforming temperature

The composition of the product gas of gasification depends on the operating conditions. Among these, temperature has a significant effect on the product composition. This effect might be connected to the effect of temperature on the chemical equilibrium of the components of the gas. Data from literature (Basu, 2010) indicated clearly that the equilibrium constants for the Boudouard reaction water–gas reaction, water–gas shift reaction and the methanation reaction were strongly dependent on temperature.

Fig. 4 shows the permanent gas composition as a function of the temperature during gasification-reforming at ER = 0.2 and S/C = 0. Increasing the temperature from 923 to 1223 K significantly increased the H₂ concentration from 17 to 38 vol.%, whereas the corresponding CO₂ concentration decreased from 27 to 6 vol.%. At low temperature, the CO concentration was almost constant at 42 vol.%. However, at high temperature, the CO concentration increased to 50 vol.%. The trend in these results suggests that high temperature gasification is recommended when high carbon monoxide and hydrogen content gases are desired.

CH₄ and other hydrocarbon concentrations were decreased by increasing the temperature. This suggests that low temperature gasification is recommended when high methane content gas is desired. This scenario is preferred when the producer gas is to be used in direct-combustion systems due to methane's high heating value (HHV).

The trend in the experimental gas composition results with respect to temperature was in agreement with that for the predicted gas composition data. A fairly good agreement between the experimental and predicted gas concentrations was noticed at high gasification temperature. Errors associated with the theoretical model assumptions at high gasification temperature were minimized due to low amounts of tar and char formation.

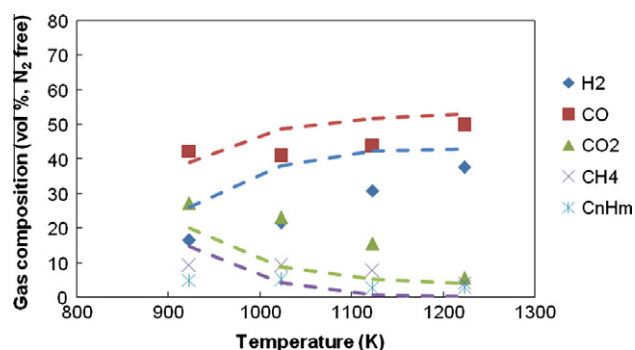


Fig. 4. Gas composition vs. temperature at ER = 0.2 and S/C = 0 (dash lines represent predicted values).

Table 1

Gasification performance at different operating conditions.

S/C (–)	ER (–)	Temperature (K)	Hydrogen yield (mol H ₂ kg ^{–1} feed)	CO efficiency (kg CO kg ^{–1} C feed) 100	HHV (MJ Nm ^{–3})
0	0.2	923	12	35	9.3
0	0.2	1023	15	31	10.4
0	0.2	1123	42	68	11.3
0	0.2	1223	54	82	12.1
1	0	1123	55	30	9.1
1	0.2	1123	59	37	7.3
1	0.3	1123	59	39	6.5
2	0.2	1123	58	26	8.4

3.1.5. Gasification efficiency

The optimum performance of a gasification process is determined based on the end-use application of the product gas. The purpose of gasification is not just energy conversion; in fact, production of chemical feedstock is also another important application. Biomass provides a good base for production of liquid transportation fuels, such as gasoline, and synthetic chemicals, such as methanol (Leduc et al., 2009). It also produces methane, which can be burned directly for energy production. There are several criteria for judging the performance of gasification processes. For example, in direct-combustion systems, the gas produced from the gasification process is sent to a nearby unit for burning such as ovens or furnaces. In such applications, the gas is fired directly while it is still hot. In this case, the product gas from the gasification process with a high HHV is required (Basu, 2010). Therefore, the hot gas efficiency defined as the ratio between the heat contained in the product gas (this include the heating values of the constituents of the gas and the sensible heat) to the heat contained in the feed serves as a good criterion for evaluating the gasifier efficiency in such application. For gasification-to-biofuels process, the CO efficiency defined as the mass of CO produced per unit mass of carbon in the feed is a good criteria for judging the economics of such processes as CO is the carbon source for the fuel synthesis (Carpenter et al., 2010). Other criteria include but not limited to: H₂/CO ratio, carbon conversion efficiency (Wu et al., 2007), gas yield and hydrogen yield (Kobayashi and Kawamoto, 2010).

Table 1 shows the mean hydrogen yield, CO efficiency and the HHV as calculated from the gas composition of each experimental result.

As it can be seen, the hydrogen yield at a temperature of 1123 K was in the range of (42–59) mol kg^{–1} feed. The production rate of H₂ was enhanced by increasing the S/C ratio. The maximum production rate of H₂ was achieved under combined steam reforming and partial oxidation conditions. Under partial oxidation conditions only, the hydrogen production was dramatically enhanced by increasing the temperature.

The CO efficiency at a temperature of 1123 K was strongly affected by the applied operating conditions. More CO was produced by increasing the ER and decreasing the S/C ratio. Similarly, under partial oxidation conditions only, the CO efficiency was dramatically enhanced by increasing the temperature.

The HHV of the product gas was in the range of (6.5–12.1) MJ Nm^{–3}. The compositions and heating values of the product gas suggest that it has a potential for application to power generation processes. Since the HHV of product gas is higher than 4.2 MJ Nm^{–3}, it is usable in gas engines (Kobayashi and Kawamoto, 2010).

4. Conclusions

We have investigated the effect of operational conditions (residence time, ER, S/C ratio and gasification temperature) on

the characteristics of product gas during cedar wood gasification. Experiments have been successfully conducted in a bench-scale gasification facility. The experimental results indicate that a synthesis gas contains around (30–50) vol.% of H₂ can be produced from cedar wood gasification. Appropriate steam and oxygen supplies are favorable for H₂ production. Higher H₂ yields, CO efficiency and HHV of the product gas can be achieved under sufficient residence times and high gasification/reforming temperatures. Increasing the S/C ratio has effectively enhanced the proportion of H₂ in the product gas. However, the presence of steam has reduced the HHV of the product gas. The presence of oxygen (ER from 0 to 0.3) can increase the H₂ yields and CO efficiency. On the other hand, excessive oxygen supply will decrease the HHV of the product gas.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.chemosphere.2012.08.029>.

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